Hot Spots in the Electronic Spectrum Renormalize the Electronic Band Structure of Actinides

Tanmoy Das,
Jian-Xin Zhu,
Matthias J. Graf, T-4;
Tomasz Durakiewicz,
John J. Joyce, MPA-CMMS

Can electrons in metallic actinides be treated as non-interacting particles or must correlations and interactions between them be taken into account, and to what extent? This is among the most fundamental questions for actinide research. The answer to this question will have far-reaching consequences. For example, it will allow us to control the functionality and thermo-physical properties of nuclear fuel. Electronic correlations, while difficult to tackle, exist in a wide class of materials, including high-temperature superconductors, itinerant-electron magnets, and heavy-fermion materials. One of the common features of these strongly correlated metallic systems is the dual nature of conduction electrons. They can behave collectively as if they had a mass many times heavier than a free electron or large magnetic moment if localized. The goal for predictive materials science is to tease out common, fundamental threads that link the localized-delocalized behavior to electronic hot spots and how they affect physical properties and functionality.

Dynamic correlation effects of strongly correlated electron systems pose a serious challenge to our theoretical understanding of the physical behavior of actinides. Strong renormalization of electronic bands and their spectral weight anomaly cannot be accounted for by the density functional theory. The interplay between $5f$ electrons with the conduction electrons pushes the system to the intermediate Coulomb-U region in which neither purely itinerant mean-field theory nor strong-coupling Anderson lattice model hold. This twilight zone of intermediate coupling gives rise to prototypical examples of strong correlations and the emergence of multiple competing phases of matter.

The metallic U-115 compounds are often contrasted with the rich phase diagram of the isostructural Pu-115 compounds $PuCoGa_5$, $PuRhGa_5$, and $PuCoIn_5$, which are superconducting with varying transition temperatures T_c. On the other hand, $UTGa_5$ (T: Ni, Pd, Pt) compounds are antiferromagnetic, while $UCoGa_5$ is paramagnetic. Due to this diversity of ground states in the U-115 and Pu-115 families and their suitability for experiments, they provide an excellent test bed for applying the self-consistent fluctuation approach and validation through experiment. By discovering the design principles for large spin and phonon fluctuations, we hope to correlate specific electronic "hot spots" in the spectral function with unique ground and excited state properties for controlled functionality of optical, thermal, and transport properties in actinides.

We start with ab initio electronic band structure calculations, which are then used as input to the self-consistent spin-fluctuation self-energy calculations [1,2]. This provides dressed electronic bands and quasiparticles with temperature dependence. The final calculations are validated by comparison with angle-resolved photoemission spectroscopy (ARPES). The ARPES data reveal strong spectral weight redistribution in the single-particle spectrum with a prominent peak-dip-hump structure, with dip around 0.5 eV in Pu-115 and U-115 materials. The separation between itinerant (peak) and incoherent (hump) states is often assigned to the duality of the $5f$ electrons. Here instead, we interpret these features in the spectrum in terms of the spin-fluctuation interaction, which creates a dip in the dressed quasiparticle spectrum due to strong scattering in the particle-hole continuum. The lost spectral weight (dip) is partially distributed to the renormalized itinerant states at the Fermi level (peak), as well as to the strongly localized incoherent states at higher energy (hump). The self-energy dressed electronic dispersions are shown in Fig. 1 for all four compounds as function of energy and momentum [1]. In all spectra the pile-up of spectral weight (hot spots) is clearly visible. At low energies all quasiparticle states are renormalized toward the Fermi level, where states are coherent and itinerant. Further away from the Fermi level, the quasiparticle states are pushed to lower binding energy. This means that the lost spectral weight is re-distributed to both lower and higher binding energies. A similar spectral weight redistribution occurs at the second peak (hump). As a result further pile-up of spectral weight occurs around 1.0-1.5 eV, creating hot spots of new quasiparticle states due to electronic correlations. Qualitatively similar behavior was also found by using the local density approximation combined with dynamical mean-field theory (LDA+DMFT) method shown in Fig. 1(e), however, with a weaker renormalization, and significantly less spectral weight near the Fermi level [3].

Our ARPES measurements in $UCoGa_5$ validated the predicted anomalous momentum and energy dependence of the electronic dispersion [2] (see

For more information contact Matthias J. Graf at graf@lanl.gov

Funding Acknowledgments
DOE, Office of Science, BES; LANL, Laboratory Directed Research and Development Program

Fig. 2). A drastic departure of the electronic states from the ab initio calculations (dashed lines) is manifest. More importantly, the associated quasiparticle width at the peak positions is significantly momentum- and energy-dependent. The energy distribution curve (EDC) of the ARPES intensity at several representative fixed momenta in Fig. 2(c), and momentum distribution curve (MDC) at several fixed energy points in Fig. 2(d) indicate that the anomaly is markedly different in energy and momentum space, which is a hallmark of electron correlations.

We conclude that the intermediate Coulomb-U coupling method of spin fluctuations provides first-principles based predictive modeling capabilities of the electronic band structure in metallic actinides. Modifying optical, thermal, and transport properties in actinides by tuning correlations will be the next step toward the ultimate goal of controlled functionality in actinide materials.

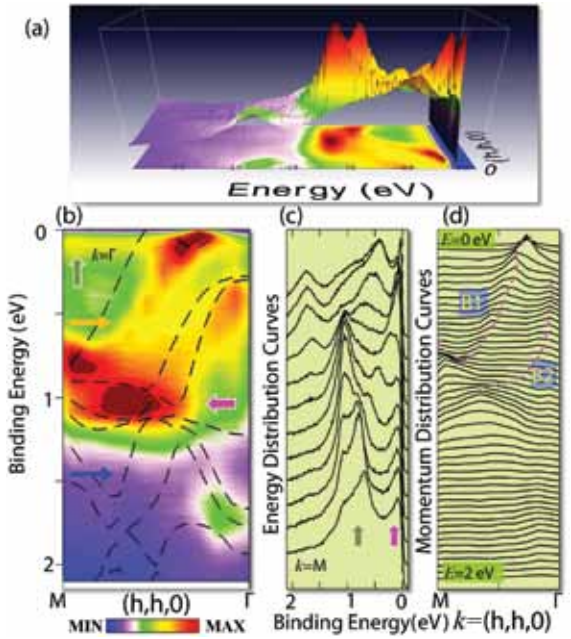

Fig. 2. Measured ARPES spectral function anomalies. (a) 3D intensity map of $UCoGa_5$ along the $M \rightarrow \Gamma$ direction (h, h, 0) in the Brillouin zone. (b) Same data plotted as 2D contour map and compared to the corresponding ab-initio electronic band structure dispersions (black dashed lines). Arrows in (b) indicate two quasi-non-dispersive energy scales of higher intensity as well as kink. (c) The EDC is plotted for fixed momenta. The curves from bottom to top are chosen for equally spaced momenta from M to Γ. Arrows have the same meaning as in panel (b). (d) The MDC for fixed binding energy. Bottom to top curves are chosen from binding energies E = 0 to 2 eV. The dashed lines are guide to the eyes for two extracted low-lying dispersions of anomalous character.

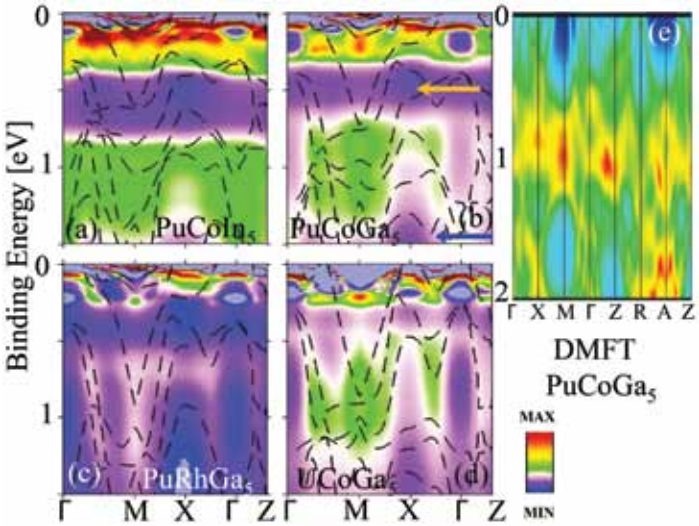

Fig. 1. (a)-(d): The self-energy renormalized angle-resolved spectral weight along high-symmetry directions in momentum space. The peak-dip-hump feature is evident in all spectra between 0 to 1 eV. Two arrows in panel (b) indicate generic dips in the spectral weight, related to peaks in the self-energy. For comparison, the hot spots in the dynamical mean-field theory (DMFT) calculations of $PuCoGa_5$ by ref [3] are shown in (e).

[1] Das, J. Zhu, X., Graf, M.J. *Phys Rev Lett* **108**, 017001 (2012).

[2] Das, T. et al., *Phys Rev X* **2**, 041012 (2012).

[3] Pourovskii, L.V. et al., *Phys Rev B* **73**, 060506(R) (2006).